BEHAVIOR OF "ONE-MODE" AND "TWO-MODE" MIXED III-V ALLOYS UNDER PRESSURE

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Résumé - L'étude par spectroscopie Raman des alliages $\text{GaAs}_{1-x}\text{P}_x$ et $\text{Ga}_{1-x}\text{In}_x\text{P}$ sous pression hydrostatique montre qu'à l'inverse de l'alliage "deux mode" $\text{GaAs}_{1-x}\text{P}_x$, les structures principales observées dans l'alliage "un mode" $\text{Ga}_{1-x}\text{In}_x\text{P}$ ne se comportent pas comme des phonons optiques de centre de zone.

Abstract - The "one mode" and "two mode" behaviors of $\text{Ga}_{1-x}\text{In}_x\text{P}$ and $\text{GaAs}_{1-x}\text{P}_x$ alloys have been studied as a function of the hydrostatic pressure by means of Raman spectroscopy. The results show that the dominant peaks do not evolve like LO and TO center zone phonons in the "one mode" system unlike the "two mode" system.

I - INTRODUCTION

The study of the vibrational properties of mixed ternary alloys have been the object of many investigations and some review papers resumed their results /1/, /2/. The main question which has not yet been satisfactorily answered is the explanation of the two types of behavior, these compounds exhibit. In III-V alloys the only "one-mode" system is $\text{Ga}_{1-x}\text{In}_x\text{P}$, at least for low x values. We present here the first results of Raman scattering experiments of $\text{Ga}_{1-x}\text{In}_x\text{P}$ under hydrostatic pressure and compare them with those of the $\text{GaP}_{1-x}\text{As}_x$ "two mode" system /3/. We have noticed some important differences between the two alloys, and this has led us to reject the assumption that the dominant peaks in the "one mode" system have still a zone center LO or TO character.

II - EXPERIMENTAL RESULTS AND DISCUSSION

Raman scattering experiments have been performed in a backward geometry, using the standard Krypton or Argon laser lines. The analyzes of the scattered light have been made through a double additive Coderg spectrometer with a resolution of 2 cm^{-1} . The high pressure was applied with a diamond anvil cell technique. This cell was designed with sapphire windows supporting the diamonds in order to increase the optical aperture of the cell. It was filled with Xenon gas which preserve the hydrostaticity of the pressure at 300 K and 80 K. The pressure was controlled by recording the emission line of different tiny ruby samples, introduced in the cell. Low temperature measurements were performed in the vacuum by cooling the cell embedded in a cold finger of a liquid nitrogen cryostat.

The spectra for different pressures are given in fig. 1 for $\text{GaAs}_{1-x}\text{P}_x$ ($x = 0.05$ and 0.11) and in fig. 2 for $\text{Ga}_{1-x}\text{In}_x\text{P}$ ($x = 0.02$ and 0.14). As one can see, the variation of the spectra is qualitatively different for the two compounds : in $\text{GaAs}_{1-x}\text{P}_x$ the dominant peaks behave independently of their relative subbands for all values of x , a behavior also encountered for the $\text{Ga}_{1-x}\text{In}_x\text{P}$ alloys with $x = 0.03$. However, for lar-

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FIG. 1 Raman spectra for different pressures of $\text{GaAs}_{1-x}\text{P}_x$ alloys for (a) $x = 0.05$ and (b) $x = 0.11$. The excitation line is 514.5 nm.

FIG. 2 Raman spectra for different pressures of $\text{Ga}_{1-x}\text{In}_x\text{P}$ alloys for (a) $x = 0.02$ and (b) $x = 0.14$. The excitation line is 514.5 nm.

ger values of x , in this latter compound, each band moves as a whole with pressure and the relative intensity of the features, inside each band, varies also significantly. We observe particularly a strong decrease of the L_1 peak versus its subband. These qualitative behaviors have been observed for all the samples we investigated in this range of concentration.

The variation of the dominant peaks that we call L_1 and T_1 with pressure is very similar to that of pure GaP for the $\text{GaAs}_{1-x}\text{P}_x$ and $\text{Ga}_{0.98}\text{In}_{0.02}\text{P}$ alloys. This similarity is also observed when plotting the difference $(\omega_{L_1} - \omega_{T_1})$ for these compounds (fig. 3). The decrease of this last quantity has been observed in all the zinc-blende type II-VI /4/ and III-V /5/ compounds. Its variation is principally quadratic. It has been interpreted as a decrease of the dynamic ionicity as a function of pressure /5/, /6/. This difference in energy is indeed that between the zone center TO and LO modes split by the coulombic interaction. The decrease of $\omega_{LO} - \omega_{TO}$ with pressure can then be attributed to the effect of pressure on the long-range forces, and appears to be characteristic of the zone center LO and TO modes exclusively. This effect is also confirmed by calculations performed on II-VI /7/ as well as III-V /8/ compounds, where the pressure coefficient of the difference between the LO and TO mode frequencies is always found to be negative around the Γ point of the Brillouin zone. In this respect, the dominant peaks in $\text{GaAs}_{1-x}\text{P}_x$ and $\text{Ga}_{0.98}\text{In}_{0.02}\text{P}$ alloys still behave as the zone center TO and LO modes.

The behavior is quite different for $\text{Ga}_{1-x}\text{In}_x\text{P}$ with $x > 0.03$. The L_1 and T_1 peaks present a higher pressure coefficient than for GaP or $\text{GaAs}_{1-x}\text{P}_x$ (fig. 4). The variation with pressure of $(\omega_{L_1} - \omega_{T_1})$ now increase linearly (fig. 3) with a -2 pressure coefficient of $(1.7 \pm 0.2) \cdot 10^{-2} \text{ cm}^{-1}/\text{kbar}$. Thus, it is derived from the preceding discussion that the experimental increase of the splitting $(\omega_{L_1} - \omega_{T_1})$ with pressure for $\text{Ga}_{1-x}\text{In}_x\text{P}$

FIG. 3 Variation of the splitting $\omega_{L_1} - \omega_{T_1}$ of $\text{Ga}_{1-x}\text{In}_x\text{P}$ and $\text{GaAs}_x\text{P}_{1-x}$ alloys as a function of the pressure.

cannot be explained assuming that these modes originated from the center of the Brillouin zone. Another singular behavior is also observed when plotting the variation of $(\omega_{L1} - \omega_{T1})$ as a function of the concentration x at atmospheric pressure: this quantity reveals a strong increase from 38 cm^{-1} for $x = 0$ to 42 cm^{-1} for $x = 0.14$ not observed in the $\text{GaAs}_{1-x}\text{P}_x$ alloys in the same range of concentration [3]. So in the "one mode" system, the main structures observed appear to originate from the activation of the density of states by the disorder, like their subbands. Physically the disappearance of the zone-center optical modes in the "one mode" system occurs when the localized density of states overlaps that of the host lattice [9]. In this situation the modes of the host lattice are significantly affected even quite far away from the impurity site, since the localized modes of the impurity can propagate inside the unperturbed lattice. Inversely for a "two mode" system like $\text{GaAs}_{1-x}\text{P}_x$ where the impurity and the host structures are separated by a "gap", such a process does not occur and the main structures will conserve their nature of center zone TO and LO modes, at least for dilute alloys.

FIG. 4 Variation of the frequencies of dominant peaks of $\text{Ga}_{1-x}\text{In}_x\text{P}$ alloys as a function of the pressure.

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